



# United States Department of the Interior

U.S. GEOLOGICAL SURVEY  
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Dear Dan,

As you requested, the document titled, "Risk and Character of Radioactive Waste at the West Lake Landfill, Bridgeton, Missouri (14 Mar 2013)" by Robert Criss, Washington University, St. Louis, Missouri, was reviewed by surface- and groundwater hydrologists of the U.S. Geological Survey Water Science Center in Rolla, Missouri. The following is a compilation of our comments on the subject document.

**G1 -Page 1, last paragraph continuing to top of page 2.—Discussion of unknown nature of leached barium sulfate wastes:**

The author is correct that samples in the 1982 U.S. Nuclear Regulatory Commission (NRC) report have barium (Ba) and sulfate ( $\text{SO}_4$ ) concentrations too small and Ba: $\text{SO}_4$  ratios too large to be primarily barium sulfate or barite ( $\text{BaSO}_4$ ); however, this discrepancy probably is the result of poor sample documentation in the NRC (1982) report, poor wording and use of industry jargon in historical documents, and ambiguity of the term "residues." The referenced report (NRC, 1982) has Ba and  $\text{SO}_4$  data for one offsite and five onsite soil samples listed in table 13. Based on the descriptions in table 13, four of the five onsite samples were surficial samples and one seems to be a subsurface sample from borehole #103. No other details are provided for the samples listed in table 13. The report text is confusing because on page 19, there is mention that six composite soil samples were collected from five auger boreholes and one sample was

collected from the Westlake treatment plant sludge and submitted for priority pollutant analysis, with a reference to table 12. The following paragraph then indicates that, “a chemical analysis of radioactive material from both areas also was performed by RMC labs and reported in table 13.” The sample numbers in table 13 (#101–#105) do not match any sample or borehole numbers in any other tables or figures in the report; however, table 4 appears to contain radiological results from analysis of several soils, but there are no sample numbers to match to table 13 and the number of samples and descriptions do not match those in table 13. If one assumes that results in table 13 are associated somehow with some of the results in table 4, there is considerable range in the radiochemical results and it is uncertain if the high radiochemical results are associated with the higher Ba concentrations.

Another possible complication is that the analytical methods for the  $\text{SO}_4$  reported in table 13 (NRC, 1982) are not stated. The reporting of  $\text{SO}_4$  in parts per million (ppm) in table 13 (NRC, 1982) is unusual because methods familiar to this reviewer (Coller and Leininger, 1955; Curry, 1996; Curry and Papp, 1996) report total S (S) content in rocks, and the S is reported in weight percent with reporting levels around 0.05 percent (500 ppm). Also, the  $\text{SO}_4$  values in table 13, even if actually in parts per million of total S, are extremely small, with several samples having reported values nearly one order of magnitude smaller than the reporting limit of 0.08 weight percent (800 ppm) in Shacklette and Boerngen (1984). However, an older method that could have been used by the NRC reports both  $\text{SO}_4$  and S contents in rocks or minerals at levels as small as a few parts per million (Vlisidis, 1966). Also, there are some methods that report water soluble  $\text{SO}_4$  but those methods may not detect  $\text{SO}_4$  locked up in insoluble  $\text{BaSO}_4$ , and thus  $\text{SO}_4$  would be under reported. Additional information is needed to adequately interpret the values listed in table 13 (NRC, 1982).

Regardless of the above, a review of the operations of the Uranium Feed Materials Process may shed some light on the disconnect the author points out between the description of the radioactive wastes placed in the landfill and the Ba and  $\text{SO}_4$  concentrations listed in table 13 of the 1982 NRC report. It is worth pointing out that NRC (1982) uses the term “leached barium sulfate residues” [underline mine for emphasis]. It is unlikely the 8,700 tons were  $\text{BaSO}_4$  but most likely were a mixture of waste that included  $\text{BaSO}_4$ . The specific reference to barium sulfate probably is because an early part of the production process called for the removal of 99 percent of the radium (Ra) by its precipitation with  $\text{BaSO}_4$ . According to a description of the production process provided in Harrington and Ruehle (1959), it appears that a large quantity of waste generated at the feed materials plant probably was from the initial acid digestion of the uranium ore and subsequent diethyl ether extraction processes. The uranium ore initially was digested in concentrated nitric acid, and then Ra was removed by the addition of barium carbonate ( $\text{BaCO}_3$ ). The  $\text{BaCO}_3$  served several purposes including neutralization of the acid mixture, removal of Ra through its association

with precipitated BaSO<sub>4</sub>, and removal of calcium (Ca) and lead (Pb) to prevent the build-up of scale on production equipment primarily from Ca and lead Pb sulfates and carbonates (Harrington and Ruehle, 1959, p. 132 and 136). The source of the SO<sub>4</sub> to precipitate BaSO<sub>4</sub> was from the uranium ore itself, but in some cases, sulfuric acid was added to the acid digestion if the original uranium ore did not have a sufficient SO<sub>4</sub> content. Thus, the Ra containing “leached barium sulfate residues” likely was a mixture of many things including excess BaCO<sub>3</sub>, BaSO<sub>4</sub>, and other sulfate and carbonate wastes with gangue and insoluble silicate minerals in the uranium ore. Thorium (Th) appears to be carried past this process in the feed materials plant because there is reference to thorium isotopes presenting a radiation hazard during the final stages of the feed materials process that involved the reduction of uranium tetrafluoride (UF<sub>4</sub>) to uranium metal (Harrington and Ruehle, 1959, p. 262).

The author’s main point perhaps is that the results in the NRC (1982) report indicate large amounts of Th-230 in some soil samples, especially compared to Ra-226, and yet generally small concentrations of Ba and SO<sub>4</sub> in the few samples listed in table 13. Although it is unknown if any of these are the same samples, it is not known if appreciable amounts of Th-230 would be present in the BaSO<sub>4</sub>-rich precipitates that should typically be enriched in Ra depending on the Ra content of the various ores processed.

## **G2- Section 2 Radiological character of the waste (p. 2–3)**

The author expends considerable effort trying to establish a representative present-day Th-230 to Ra-226 ratio using sparse and variable ratio data from NRC (1982) and NRC (1988). NRC (1982) does present tables of surface soil activity collected on a 10 meter (m) by 10 m grid pattern across Area 1 and Area 2, and activity in selected boreholes using a NaI(Tl) detector [Bi-214 (bismuth-214) was used as a surrogate for Ra-226]; however, only 12 samples were submitted for analysis of Th-230 activity (NRC, 1982, table 4) and the sample locations are not indicated clearly. On the other hand, the Engineering Management Support, Inc. remedial investigation report (EMSI, 2000) contains data from a comprehensive radiological survey of surface and subsurface soil across Area 1 (tables B-1, B-3, and B-5), Area 2 (tables B-2, B-4, and B-6), and the Ford property (tables B-7, B-8, and B-9). These data offer the possibility of constructing a detailed distribution map of present-day Th-230:Ra-226 activity ratios. A quick scan of these data indicates a few ratios in the range of 30:1 to 100:1, but the majority of ratios are well below 30:1.

The author is correct in the analysis that with Th-230:Ra-226 ratios above 1.0, the maximum increase in Ra-226 is not reached for about 9,000 years, when Ra-226 ingrowth activity comes into secular equilibrium with Th-230. The increase in total activity is substantial, for example, starting with an initial Th-230 of 52,000 picocuries per gram (pCi/gm) and Ra-226 of 3,900 pCi/gm (NRC, 1982, table 4, sample 11J), the total alpha activity at about 9,000 years would be nearly 200,000 pCi/gm [Th-230+Ra-226+Rn-222

(radon-222)+Po-218 (polonium-218)]. A full analysis of the data in the EMSI (2000) report would be appropriate.

### **G3-Section 4 Hydrologic and Geologic Risk Factors**

*G3A – Page 5 second paragraph, beginning with second sentence, “Such simplistic statements ignore persuasive evidence that flood levels on the lower Missouri and Mississippi Rivers have been increasing with time (Criss and Shock, 2001)”.*

This broad statement is an oversimplification and the subsequent discussion in the paragraph seems to imply that there is some arbitrary and unexplained increase in flood levels with time. There are logical and documented causes for increased flood levels, most of which is captured in the title of the referenced Criss and Shock (2001) report, "Flood enhancement through flood control." The general statement that flood levels have increased with time on the lower Missouri and Mississippi Rivers is not altogether unfounded. The author has justifiably raised awareness of one of the consequences of flood plain development. An independent detailed examination of streamflow records on the middle Mississippi River in the St. Louis area by the U.S. Geological Survey (USGS) (Huizinga, 2009) that was not referenced by the author provides additional detail and explanation for changes in stage-discharge relations and rating curves.

The primary argument seems to hinge on the definition of the "500-year" flood level of the Earth City levee. The reviewers understand that the "500-year" flood actually is the probabilistic definition wherein a flood has a 1 in 500 (0.2 percent) chance of occurring in any given year, and therefore can occur more frequently than "once every 500 years." The statement that there have been several "100-year" and "500-year" floods in recent years is unsubstantiated, with the primary evidence being the flooding in the middle Missouri River in 2011, which was a reservoir-induced flood (Huizinga, 2012). The main point, however, is that it is important to realize that any given recurrence interval, or more accurately, any annual exceedance probability, is based on analysis of past data, and continuously is refined as additional data are collected. This continual refinement is the very nature of the flood science. Also, any given flood can be overtopped (that is, there can always be the "1000-year" flood, even if we had the "500-year" flood last year, or even last month). Thus, flood peak statistics can be constantly revised with every new peak.

*G3B – Second paragraph, third sentence from the end, beginning with, “Statistical analysis of actual flood records shows that the recurrence statistics promulgated by the US Army Corps of Engineers (USACE, 2004) typically have less than a 1% chance of being realistic.”*

It is unclear what the author's evidence is for the statement that the USACE's recurrence statistics, "have less than a 1% chance of being realistic." Every probabilistic recurrence interval has error bars that increase with the magnitude of the event because there is much less data available. To be accurate, this statement needs to be made in the context of error and a specific definition offered for the term "realistic." If a predicted flood peak level for a 1.0-percent event at a site is 30.25 feet (ft) based on data through 2003, but analysis of additional data collected through 2013 results in a revised 1.0-percent level to 30.30 feet, or working backwards with the new streamflow data indicates that the 30.25-ft level statistically is really a 1.02-percent event, is that considered incorrect? If the error around the 1.0-percent event is 10%, then the original value, although revised, is not incorrect.

#### **G4 – Section 5, Groundwater Contamination**

Page 6, paragraph 1, third sentence, "*The water table in the alluvial aquifer is known to rapidly respond to the river stage as well as to the delivery of recent precipitation, with groundwater rapidly moving either toward or away from the river, depending on the river stage (e.g., Emmett and Jeffrey, 1968; Grannemann and Sharp, 1979; Criss and Criss, 2012).*" [underline mine for emphasis]

The underlined part of the statement is incorrect. A rapid change in water level measured in an alluvial aquifer well associated with changes in river stage does not indicate rapid movement of the water itself, but the rapid propagation of a pressure head. This is a common misconception and given the author's background and discussion on the following page, probably an unintended misstatement. As the author indicates later, the hydraulic conductivity of the alluvial aquifer can be large. The large hydraulic conductivity leads to the rapid propagation of head changes in alluvial wells in response to river changes, but not the actual movement of the water within the aquifer. This phenomenon is discussed in more detail in comments on the following page.

Although Granneman and Sharp (1979) describe the various complexities of general types of head response in the alluvium to river stage, and show how the system could be simplified for digital modeling, they do not indicate that flow rates are rapid. In the last 20 years, the USGS has done extensive modeling of the Missouri River alluvial aquifer in the Kansas City area (Kelly, 1995, 1996a, 1996b, 2002, 2010) ; however, the author does not reference this extensive recent literature that relates directly to the issue of rates of groundwater movement in the alluvial aquifer the author raises. The recent studies include traveltime estimates and flow path tracking that have increased understanding of groundwater flow in the alluvial aquifer beyond the tools available to Granneman and Sharp (1979).

## **G5 Section 6, Groundwater migration**

*G5A – Section 6, third paragraph, second sentence beginning with, “Contrary to their claims (EMSI 2012; p. iii and p. 9), the potentiometric surface map in this report (Fig. 2; EMSI 2012) clearly shows that this piezometer is far downgradient, not “upgradient”, of the water table in Area 1, so that the radiological contamination has migrated radially away from Area 1, as well as downward into the Mississippian bedrock aquifer.”*

The author is correct that the large Ra-226 in PZ-101-SS is consistent with migration of Ra-226 into the bedrock, possibly from Area 1. The sample also contained concentrations of sodium (27 mg/L [milligrams per liter]), chloride (180 mg/L), and bromide (1.1 mg/L) that may indicate landfill leachate. Additional data are needed, especially on radionuclides from background wells in the vicinity of the site. A review of groundwater level data in the EMSI (2012) report, and subsequent data collected during April 2013, indicates that there does not appear to be a large groundwater “mound” beneath Area 1 that would drive flow counter to the more natural expected flow path that is to the north or northwest.

Leachate migration into the Mississippian bedrock aquifer beneath the alluvium probably would have to be density driven under natural hydraulic conditions, because the alluvium is a discharge area for regional groundwater flow; however, pumping to dewater the large landfill in the former quarry area south of Area 1 may have reversed the natural groundwater direction in some areas and potentially caused leachate to move counter to the natural alluvial gradient. Besides PZ-101-SS, there are several detections of radionuclides in piezometers south and east of the quarry that cannot be explained by migration from Area 1 under the hydraulic gradient shown in EMSI (2012, fig. 2). Even in the absence of a pumping-induced drawdown, some lateral migration of leachate within the landfill debris might be expected because of the discontinuous nature of the landfill material. The potential for offsite migration of leachate will not be understood or assessed fully unless groundwater levels are monitored (some continuously) and groundwater samples are collected from the expected higher-permeability zone at the base of the alluvial aquifer offsite around the northern end of the landfill.

*G5B – page 6, last paragraph, sentence beginning, “However, these measurements are not typical as they were made in late July, 2012, in the middle of a protracted drought.”*

The author implies that EMSI (2012) reported gradients were too flat and that gradients beneath the landfill are much larger. As evidence for this, the author references a statement in NRC (1988, p. 6), “Water levels recorded between November 1983 and March 1984 in monitoring wells at the landfill, indicate a

groundwater gradient of 0.005 flowing in a N 30°W direction beneath the northern portion of the landfill.” In the absence of data to verify the gradient reported, the 0.005 value, although nearly 10 times the value listed in EMSI (2012), is still a small value.

An average gradient across the site was estimated using well pairs I-68 and D-81 as upgradient wells and well pairs I-65 and D-93 as downgradient wells. On July 30, 2012, the horizontal gradient was 0.0007 and on April 3, 2013, the average gradient from these same pairs was also 0.0007. The daily mean altitude of the Missouri River at the USGS St. Charles streamgage (station 06935965) on these two dates was 422.68 ft and 425.64 ft. Given the river at the streamgage is about 9,400 ft from the site, hydraulic gradients across the entire alluvium from the site to the river streamgage were calculated to be 0.0008 and 0.0002 for the two dates. The average gradient using well pairs at the site is consistent with the overall gradient in the alluvium. Using a range of hydraulic conductivity (K) values from 85 to 400 ft/day (feet per day) and an average porosity of 0.15 to 0.30 for silty-sand and sand sediments, and the gradient of 0.0007 ft/day, the estimated groundwater flow rate ranges from about 0.2 to 1.8 ft/day. Flow rates through silt and clay layers, which are common, in the Missouri River alluvium are much slower (Grannemann and Sharp, 1979).

If landfill leachate and radionuclides migrated into the alluvial aquifer, and have been migrating from the landfill at the estimated flow rate for the past 40 years, the leachate potentially would have moved thousands of feet and easily reached the Missouri River. These simple 1-D calculations, although useful to gain perspective, are not accurate. The actual movement of any leachate that migrates into the alluvial aquifer is affected by environmental conditions such as leachate-sediment reactions, heterogeneity in aquifer properties, leachate density, groundwater gradient variations in response to aquifer-river interactions, presence of nearby alluvial pumping wells, and other factors.

*G5C – page 7, first complete paragraph beginning with, “EMSI (2012, p. 7) similarly underestimates the hydraulic conductivity of the alluvial aquifer, stating that their measurements indicate that it is only 8.5 to 85 ft/day.”*

Care must be taken when evaluating hydraulic conductivity of the alluvial aquifer. It is likely that all the values listed here by the author, including those from EMSI (2012), may be correct. The hydraulic conductivity of the alluvial aquifer is related to the grain size of the sediments, which exhibit an exponential increase with depth (Granneman and Sharp, 1979). Grain size also typically tends to be finer near the valley walls where the alluvium often is thinner. The hydraulic conductivity increases as the grain size increases

and, therefore, with depth, reaches a maximum in the deepest parts of the alluvium. The 400-ft/day values listed by Emmett and Jeffery (1968) reflect the highly permeable coarse sands and gravels in the lower part of the alluvium targeted by public supply and irrigation wells. In contrast, hydraulic conductivity values measured in the Missouri River alluvium at the Riverfront Site in New Haven, Missouri, ranged from 10 to 22 ft/day in sands and silty sands in the upper part (25–50 ft deep) of the alluvium near the edge of the alluvial valley (EPA, 2003). Kelly (1995) states that reported conductivity values for the alluvial aquifer in the Kansas City area range from 126 to 325 meters per day (414 to 1,066 ft/day). These values are for the deeper coarse grained parts of the aquifer. Values for the shallow parts of the alluvial aquifer in the Kansas City area can be as low as 0.1 meters per day (0.3 ft/day) in clay-rich zones.

*G5D – page 7, second paragraph, third sentence, “Instead, the NRC data indicate that the velocity would be more than 100x faster than EMSI’s upper limit.”*

Groundwater levels in the alluvial aquifer are in a constant state of flux as levels in the alluvium respond to changes in river stage, especially in closer proximity to the river. As these head changes work their way through the aquifer, groundwater can even flow away from the river for brief intervals. The rapid changes in hydraulic head do not indicate a rapid flow of groundwater through the aquifer. The referenced alluvial aquifer hydraulic gradient of 0.005 in the NRC (1988) report seems more atypical than the reported 0.0004 (EMSI, 2012) and average of 0.0007 estimated in comment G5B. The question also arises as to whether or not both gradient estimates were based on measurements in the same wells.

Calculations of estimated groundwater flow rates must be done in context with a conceptual site model and specific units or intervals. Using hydraulic conductivity values for one area of the alluvium and hydraulic gradients from another can result in erroneous estimates. Likewise, care should be taken with the author’s “100x faster” velocity estimate because that value also may be determined from a mix of data from different areas and depths in the alluvium.

*G5E – page 7 second paragraph, last sentence, “It should also be mentioned that these so-calculated “Darcy velocities” are about 4x slower than the actual microscopic velocity of the groundwater, because the real groundwater velocity also depends on the alluvium porosity.”*

The author is correct that the reported range of groundwater flow velocity values listed in EMSI (2012) of 0.0034 to 0.034 ft/day (1.2 to 12 ft/year [feet per year]) are too low. The values in EMSI (2012)



were not adjusted for effective porosity of the aquifer. Assuming a silty-sand to sand effective porosity value of 0.15 to 0.30, the EMSI (2012) flow estimates should be about 0.01 to 0.23 ft/day (4 to 84 ft/yr).

#### **G6 Section 7, Background Radiation Levels**

The author is correct that the Szabo and others (2012) reference in the EMSI (2012) report is not appropriate because bedrock beneath the West Lake Landfill is part of the Mississippian-age Springfield Plateau aquifer and not the deeper and underlying Cambrian-Ordovician age rocks of the Ozark aquifer system that is discussed in Szabo and others (2012). Even if Szabo and others (2012) was applicable, the nearest sample location shown on figure 1 of their report is in southwestern Missouri, nearly 200 miles from the West Lake Landfill. The EMSI (2012) report initially appears to present the Szabo and others (2012) data in the context that elevated radionuclides can be present in some regional aquifer systems, but then clearly takes this concept out of context in statements on page 13, “The median level of 5.9 pCi/L reported by Szabo et al. (2012) for the MCOO aquifer system is higher than the median total concentration of combined Radium-226 and Radium-228 found in either bedrock or alluvial monitoring wells at the Site (Table 9). This indicates that the levels of radium detected in the monitoring wells reflect natural occurrences of radium.” The author is correct in his criticism of the EMSI (2012) report. To conclude that background Ra-226 and Ra-228 levels in the Mississippian-age bedrock beneath the Westlake Landfill can be determined by data published in Szabo and others (2012) is erroneous.

Perhaps some clarification is needed because Szabo and others (2012) combines NAWQA (National Water Quality Assessment) data from two different regional aquifer systems into what is called the MCOO (Mid-continent and Ozark Plateau Cambro-Ordovician dolostones). The Missouri River is the boundary between the Cambrian-Ordovician aquifer system to the north (Missouri, Iowa, Illinois, and Wisconsin) and the Ozark Plateaus aquifer system to the south (mostly southern Missouri with parts of Arkansas, Kansas, and Oklahoma). The names are different, but the rock strata are similar. The referenced report (Szabo and others, 2012) was a regional assessment of radionuclides in groundwater covering the central and eastern United States. Szabo and others (2012) combined data collected by the NAWQA program from the formal Cambrian-Ordovician aquifer system to the north and the similar age rocks of the Ozark aquifer of the Ozark Plateaus aquifer system to the south. In Szabo and others (2012), figure 1 shows several sample locations in southwestern Missouri; however, samples from these locations were not analyzed for Ra-226 or Ra-228 but for total alpha emitting isotopes from radium. Perhaps a better reference is Focazio and others (2001), who determined concentrations of Ra and other radionuclides in 10 public-supply wells in Missouri; however, like Szabo and others (2012), none of the wells sampled by Focazio and others

(2001) were in the vicinity of the West Lake Landfill. Concentrations of Ra-226 reported by Focazio and others (2001) ranged from 0.53 to 8.96 pCi/L from wells more than 650 ft deep.

The USGS National Water Information System (NWIS) database has 43 groundwater samples in Missouri (bedrock or alluvium) that were analyzed for Ra-226 and 39 samples that were analyzed for Ra-228 (<http://waterdata.usgs.gov/mo/nwis/qw/>). The maximum Ra-226 concentration in these samples is 0.73 pCi/L and the maximum Ra-228 concentration is 2.0 pCi/L. Of these samples, there are only three in the Mississippian-age bedrock and these are from three isolated monitoring well samples at the Weldon Spring Site and have a maximum Ra-226 concentration of 0.57 pCi/L and Ra-228 concentration of less than or equal to 1 pCi/L. Additional information is needed on background concentrations of radionuclides in the vicinity of the site in both the alluvial aquifer and the Mississippian-age bedrock aquifer.

#### **G7 Section 8, Assessment and Recommendations**

- Bullet 1            Although the author may be correct that the majority of the radioactive material (RIM) is not barium sulfate or barite, in reports on the site there are only a few poorly documented chemical analyses compared to the hundreds of radiological measurements, so it is difficult to substantiate the author's statement that longstanding assertions indicating that barium sulfate is an important component of the radioactive wastes at the site is somehow contrary to chemical and physical data. The previous reports do not use the term "leached barium sulfate" that the author does, but use the term "leached barium sulfate residues". This may be an important distinction and a careful review of the feed materials process may provide some information on the general nature of what the "residues" may have been.
  
- Bullet 2            The author is correct that ingrowth of Ra-226 and daughters from Th-230 will increase the activity of the RIM for about 9,000 years.
  
- Bullets 5&6        Initial data from the quarterly monitoring indicate elevated levels of radionuclides in groundwater such as Ra-226 in monitoring well PZ-101-SS and others, as the author has pointed out.
  
- Bullet 7            The author is correct that there is a sparse amount of radionuclide data from the alluvial aquifer and Mississippian-age bedrock aquifer in the vicinity of the site that makes determination of background levels difficult and that EMSI (2012) does not make a valid case for background values.

Bullet 8            The author is correct that to determine if landfill leachate and radionuclides have migrated some distance offsite and down the presumed direction of groundwater flow in the alluvium, additional offsite monitoring wells, particularly north and northwest of Areas 1 and 2, would be needed. Stable isotopes are a powerful tool in groundwater studies and could provide additional information; however, a careful examination of other elements that the author mentions (including major and trace inorganic constituents such as sodium, chloride, boron, and others), which are commonly associated with municipal landfill leachate at reporting levels sufficiently low to capture ambient levels, is needed to provide additional information on migration of leachate at the site.

Please contact me if you have any questions at 573-308-3678 or email at [jschu@usgs.gov](mailto:jschu@usgs.gov).

Best Regards,

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